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Coaxial HgI excimer lamps

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Abstract. **The emission of coaxial HgI excimer lamps pumped by a repetitively pulsed barrier discharge is experimentally studied. The stable operation of the excimer lamps was demonstrated at pump-pulse repetition rates from 0.5 to 12 kHz, and the average emission power attained of 0.6 W at 444 nm. It was found that upon an addition of 0.8 % of xenon to the mixture of helium and mercury diiodide, the pulse and average emission powers increased by 30 %. The emission power reduced by 5 % after 2.5×10^6 pulses. An interpretation of the results of optimising the excimer lamp characteristics is given.**

Keywords: *excimer lamp, barrier discharge, gas-discharge plasma, mixture components, mercury diiodide, inert gases.*

1. Introduction

Interest in excimer sources of spontaneous visible, UV, and VUV radiations (excimer lamps) is due to a number of facts related to the practical applications of such lamps [1 – 6]. For instance, many applications require only an incoherent high-power source with a large area (more than 100 cm^2) emitting in a relatively broad wavelength range. In addition, excimer lamps are simple to fabricate and noncritical to the pump power and the geometry of the active region. This makes promising the use of the emission of such lamps in the visible range in agricultural physics for efficient excitation of chlorophyll molecules, thus increasing the rate of plant growth, as well as for pumping dye lasers, and in medicine and biotechnology.

So far several excimer sources of spontaneous visible light have been built and studied [6 – 8]. In these sources, barrier or other types of discharge are used to excite the working mixture. The majority of studies in this field deal with the excimer molecule HgBr* (mercurous bromide).

In this paper, we study experimentally the effect of composition of the working mixture, the voltage applied to the excimer lamp, and the pulse repetition rate on the spectral, energy, and temporal characteristics of the emission of a coaxial HgI excimer lamp.

2. Experimental

We studied the emission characteristics in $HgI_2 - He$ and $HgI_2 - N_2(Xe) - He$ mixtures in cylindrical excimer lamps of two types providing the pumping of the working mixtures by a repetitively pulsed barrier discharge. Fig. 1 depicts the design of the coaxial excimer lamps. The lamps were fabricated from quartz tubes 20 cm long and 34 mm in diameter. In one lamp (Fig. 1a) a tungsten electrode (*4*) of a circular cross section with diameter of 4 mm is directed along the axis, while in the other (Fig. 1b) two such electrodes are symmetrically displaced with respect to the axis and are separated by a distance of 20 mm. The second electrode (*3*) in each lamp is perforated (with a radiation transmission coefficient of 72 %) and is located on the external surface of the quartz tube along its entire length. The electrodes DRT-240 of quartz lamps welded into the end faces of coaxial lamps were used as leads (*6*) providing a contact with the main electrode(s) (*4*). The opposite end face had an outlet (*1*) made of quartz glass with a capillary 1 mm in diameter, which was used to diminish the outflow of mercury diiodide vapour from the lamp into the evacuation system.

The excimer lamps were excited by a nanosecond pulse oscillator (Fig. 2). A TGI2-130/10 gas-filled triode was used as the commutator C, and a storage capacitor SC consisted

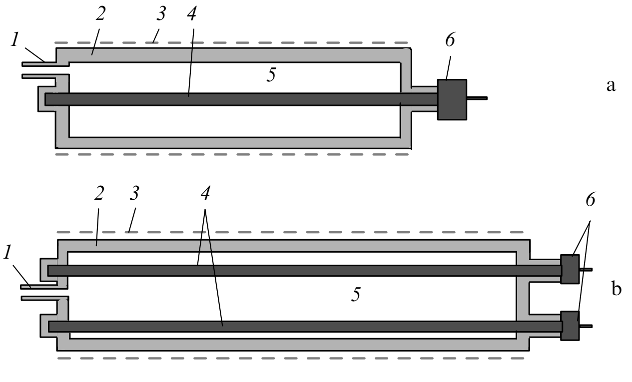

Figure 1. Design of coaxial excimer lamps: (*1*) outlet for evacuating the lamp and gas admission; (*2*) quartz tube; (*3*) perforated electrode; (*4*) electrode; (*5*) discharge region; (*6*) high-voltage lead.

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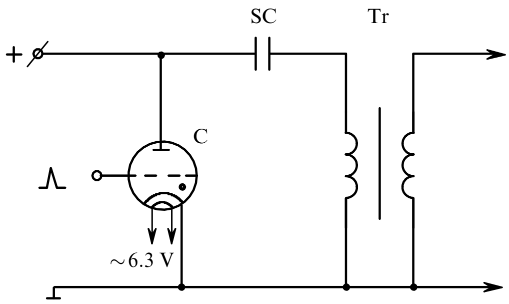

Figure 2. Electrical circuit of the pump oscillator: (C) commutator; (SC) storage capacitor; (Tr) step-up transformer.

of low-inductance KVI-3 capacitors. The storage capacitor SC was recharged through the primary winding of a step-up transformer Tr with a 1:3 transformation ratio. In the process of the experiments the voltage across the capacitor was varied from 4 to 9 kV, the pulse repetition rate, from 0.5 to 12 kHz, and the capacitance of the storage capacitor was 1.36 or 3.4 nF. The voltage and discharge current pulses of the excimer lamps were recorded with an S8-2 dual-beam oscilloscope after passing through a voltage divider and a calibration Rogowski loop, respectively. The amplitude–temporal characteristics of the emission in the 400–450 nm range selected by a SZS-8 light filter were measured with a FEK-22SPU photocell, whose output signal was recorded with the S8-2 oscilloscope. A diaphragm with an area of 1 cm^2 was placed in front of the photocell. The average emission power in this range was measured with a Kvarts-01 power meter.

We studied the emission spectra of the gas-discharge plasma by focusing the plasma emission propagating through a diaphragm of area 1 cm^2 by a lens on the entrance slit of a ZMR-3 prism monochromator. The optical signal was detected with a FEU-79 photomultiplier, amplified with a U1-2 amplifier, and recorded with a KSP-4 chart recorder (the detection system was similar to that used in paper [6]). The reciprocal linear dispersion of the ZMR-3 monochromator was 44 Å mm^{-1} at 434 nm. The emission spectra were recorded using the monochromator slits of 0.1 mm, while in the measurements of the integrated characteristics (the dependence of the spectral band emission intensities on the mixture composition) the slits were 1.5 mm. The detection system was calibrated using an SI8-200 standard ribbon filament tungsten lamp with the filament temperature of 2173 K.

The working mixtures were prepared directly in a gas-discharge cell by filling it with the heavy inert gas (xenon) or nitrogen and then with the light buffer gas (helium). Sixty milligrams of mercury diiodide were loaded into the excimer lamp before the experiment. The lamp was degassed by heating at 50 °C and evacuating for two hours. The partial pressure of the HgI_2 vapour in the working mixtures was produced by heating the mixture during the dissipation of energy of the high-frequency discharge and was determined from the temperature of the coldest part of the lamp by linearly interpolating the data taken from Ref. [9]. In contrast to our earlier study [6], the excimer lamps were thermally insulated to increase their heating temperature and, thereby, to increase the partial pressure of mercury diiodide vapour to the optimal value [7]. The thermal insulation was achieved using a fluoroplastic film of thickness ~0.2 mm covering the excimer lamp. The partial pressures of the gases were measured by a standard membrane vacuum gauge or manometer.

3. Results and discussion

3.1 Spectral and integrated characteristics of emission

The emission spectra of the HgI excimer lamps were studied in HgI_2–He and HgI_2–N_2(Xe)–He working mixtures that differ in component composition in the following partial pressure ranges: 100–180 kPa for helium, 0.5–2.5 kPa for nitrogen, 0.5–7 kPa for xenon, and up to 65 Pa for mercury diiodide. The voltage, current, and pump-pulse repetition rate were 12–27 kV, 350 A, and 0.5–2 kHz, respectively.

The characteristic emission spectrum of the HgI excimer lamp with two inner electrodes (see Fig. 1b) for the HgI_2–He working medium at $f = 2$ kHz is depicted in Fig. 3a and that for the HgI_2–Xe–He mixture at $f = 0.5$ kHz, in Fig. 3b. In the first case, only emission corresponding to vibronic bands of the $B^2\Sigma^+_{1/2} \rightarrow X^2\Sigma^+_{1/2}(v' = 0-5, v'' = 9-19)$ transition in HgI* molecules was observed [10, 11] with a maximum at $\lambda = 444$ nm, the emission band intensity sharply increasing from the long-wavelength side, and gradually decreasing from the short-wavelength side (Fig. 3a). Similar spectra are observed for mixtures with molecular nitrogen and xenon. For the mixture with xenon at reduced pump-pulse repetition rates (500 Hz, Fig. 3b), in addition to the system of bands of the $B \rightarrow X$ transition, the xenon line at $\lambda = 823$ nm, the $6s[3/2]^0_2 - 6p[3/2]_2$ transition, and weakly resolved peaks corresponding to the xenon lines $6s[3/2]^0_2 - 7p[3/2]_2$ (462 nm) and $6s[3/2]^0_2 - 7p[5/2]_2$ (467 nm) were observed. A characteristic feature of the mixture with nitrogen at $f = 500$ Hz is the presence of the second positive system (the $C^3\Pi_u \rightarrow B^3\Pi_g$ transition at 337, 357, and 380 nm) and helium lines (the $2p\,^3P^0_{2,1,0} - 3s\,^3S_1$ and $2p\,^3P^0_0 - 3d\,^3D_{3,2,1}$ transitions at 706 nm and 587 nm), as is the case with the HgBr/HgCl excimer lamp [6]. The emission spectra were identified using the data from handbooks [11, 12].

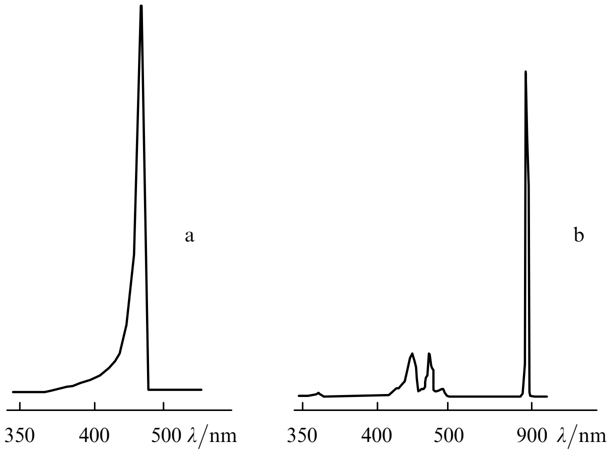

Figure 3. Emission spectra of the HgI excimer lamp with two inner electrodes for mixtures HgI_2 : He = 2.6 Pa : 162 kPa, $f = 2$ kHz (a) and HgI_2 : Xe = 0.08 Pa : 1 kPa : 161 kPa, $f = 500$ Hz (b) at the voltage applied to the excimer lamp equal to 22.5 kV and the current equal to 350 A.

The results of our studies of the integrated characteristics (the dependence of the average radiation power on the partial pressures of the buffer gas (helium), molecular nitrogen, and xenon) and the time of operation of HgI excimer lamps on one portion of the working mixture under optimal partial pressures of the mixture components are shown in Figs 4–6. The maximum radiation power is achieved at a helium partial pressure in the 160–180 kPa range, a nitrogen partial pressure of 1.5 kPa, and a xenon partial pressure of 1 kPa. The drop in radiation power after 2.5×10^6 pulses was no more than 5 % for all mixtures studied.

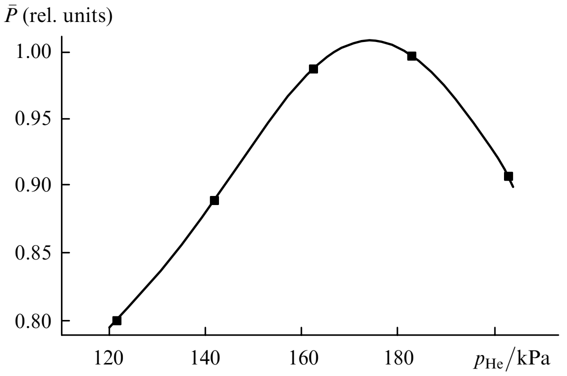

Figure 4. Dependence of the average emission power on the partial pressure of helium at $f = 2$ kHz for the voltage applied to the excimer lamp equal to 22.5 kV.

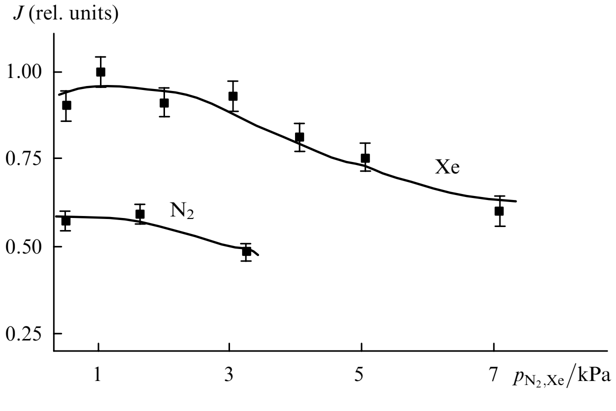

Figure 5. Dependences of the emission intensity of HgI^* molecules on the partial pressures of xenon and nitrogen for the total pressure of the mixture equal to 162 kPa.

3.2 Temporal and energy characteristics of emission

The temporal and energy characteristics of the radiation emitted by HgI excimer lamps are presented in Figs 7–9. The lamp operation was found to be stable within the parameter ranges specified in these figures. Fig. 7 shows the oscillograms of the discharge current and emission pulses in mixtures of mercury diiodide with helium, xenon, and nitrogen for the excimer lamp with one inner electrode (Fig. 1a) under experimental conditions that are optimal with respect to emission power. The error and reproducibility of the results of oscilloscope measurements were 10 % and 90 %, respectively. Current pulses had opposite polarities, an amplitude of about 350 A, and a duration of about 150 ns; their complex shape is caused by the recharge of the dielectric–plasma circuit. For the HgI_2–He, HgI_2–Xe–He, and HgI_2–N_2–He mixtures, the pulse durations at half-maximum were ~ 150, ~ 120, and ~ 150 ns, respectively. Note that the amplitude of the second pulse in the HgI_2–Xe–He mixture is smaller than that of the first, while in the HgI_2–He and HgI_2–N_2–He mixtures the situation is the opposite.

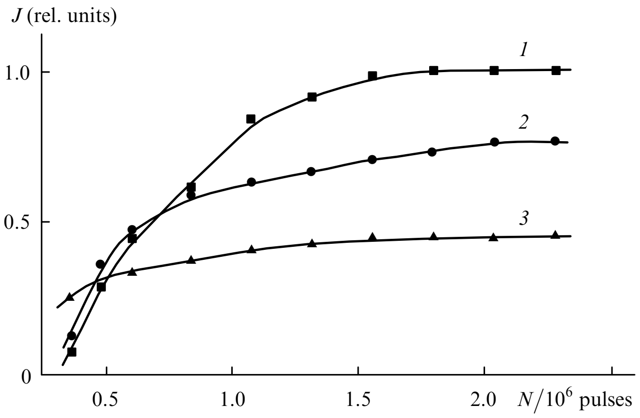

Figure 6. Dependences of the emission intensity on the total number of pulses for mixtures $HgI_2 : Xe : He = 30$ Pa : 1 kPa : 161 kPa (*1*), $HgI_2 : He = 30$ Pa : 162 kPa (*2*), and $HgI_2 : N_2 : He = 30$ Pa : 1 kPa : 161 kPa (*3*).

The maximum pulse radiation power of 130 W was achieved in a mixture of mercury diiodide with helium and xenon. In the excimer lamp with two inner electrodes (in contrast to the lamp with one electrode), the radiation power was distributed nonuniformly over the surface, namely, the radiation power in the plane containing the two electrodes was higher than that in the direction perpendicular to this plane by a factor of 1.5. The average radiation power increased linearly with increasing the pulse repetition rate up to 12 kHz (Fig. 8).

An increase in the capacitance C of the storage capacitor from 1.36 to 3.4 nF results in an increase in the average radiation power by a factor of 4.7 (Fig. 9). This is caused by the better matching between the output resistance of the oscillator and the load (lamp). The highest average power (0.6 W) was obtained in the $HgI_2 : Xe : He$ mixture with a 1 : 15 : 2388 composition at a total pressure of 162 kPa and a pump-pulse repetition rate of 12 kHz ($C = 3.4$ nF).

The discharge in all the mixtures resembled the typical barrier discharge [2, 7, 13, 14]. As the pump-pulse repetition rate increased, the radiation intensity of a homogeneous discharge also increased, while the radiation intensity in the filamentary channels decreased. The thickness of the discharge region and the discharge length were 13 mm and 20 cm, respectively.

The changes in the emission spectrum of the HgI excimer lamp at a pulse repetition rate of 500 Hz (see Fig. 3b) are caused by a decrease in the temperature of the working mixture, resulting in a drop in the partial pressure of the saturated mercury diiodide vapour and, respectively, in a decrease in the concentration of HgI_2 molecules. This, in turn, reduces the intensity of the 444-nm emission band [7]. The xenon and helium lines and the nitrogen bands can be observed by increasing the sensitivity of the detection system.

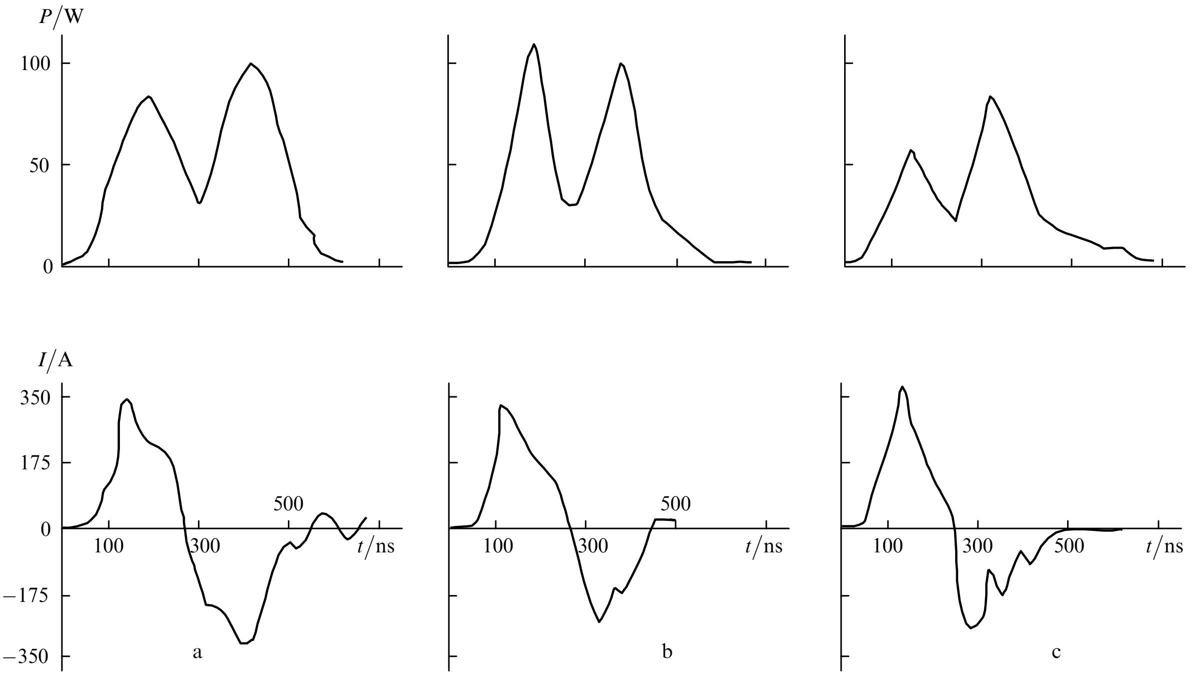

Figure 7. Oscillograms of pulses of the emission power P and discharge current I in mixtures $HgI_2 : He = 0.24\ Pa : 162\ kPa$ (a), $HgI_2 : Xe : He = 0.24\ Pa : 1\ kPa : 161\ kPa$ (b), and $HgI_2 : N_2 : He = 0.24\ Pa : 0.5\ kPa : 161.5\ kPa$ (c) at $C = 1.36$ nF, $U = 22.5$ kV, and $f = 2$ kHz.

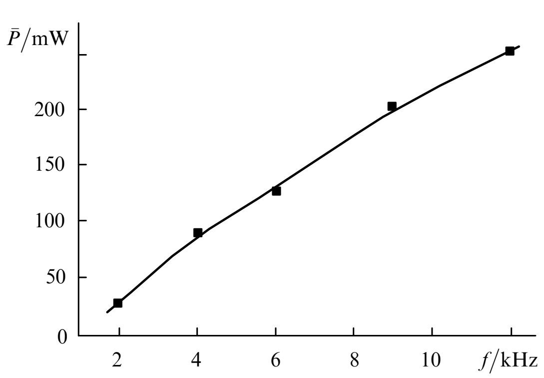

Figure 8. Dependences of the average emission power on the pulse repetition rate for the $HgI_2 - He$ mixture for a total mixture pressure of 162 kPa with $U = 22.5$ kV and $C = 1.36$ nF.

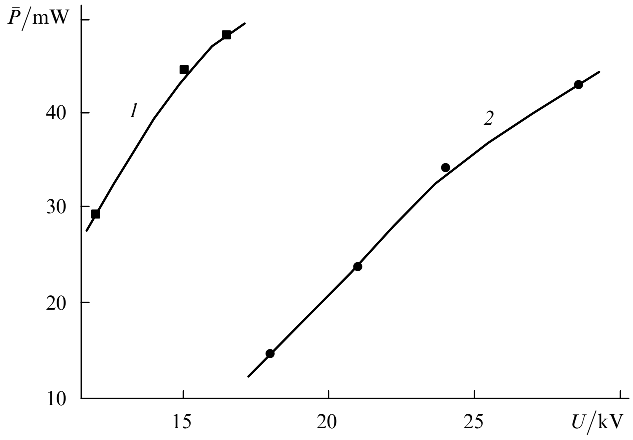

Figure 9. Dependences of the average emission power on the voltage applied to an excimer lamp operating on the $HgI_2 - He$ mixture for $C = 3.4$ (*1*) and 1.36 nF (*2*); the total mixture pressure is 162 kPa, and $f = 2$ kHz.

The existence of an optimal partial pressure of the buffer gas (helium; see Fig. 4) is related to the fraction of the discharge energy spent for heating the working mixture. Our earlier research [7] has shown that upon producing the partial pressure of mercury diiodide vapour by heating the excimer lamp with an external electric heater there exists the partial pressure that is optimal with respect to the radiation power emitted by the HgI^* molecules. In our case (for pump-pulse repetition rates of about ~ 2 kHz), the optimal range of partial pressures of helium ensures such an energy contribution to the discharge that the heating of the excimer lamp to a temperature of about ~ 130 °C produces a partial pressure of the saturated vapours (65 Pa) close to the optimal one [7].

The addition of nitrogen [the lower curve in Fig. 5 and curve (*3*) in Fig. 6] results in a decrease in the radiation power compared to that emitted by a lamp with the two-component mixture because the discharge energy is spent for additional vibrational excitation of molecular nitrogen [15].

The increase in the radiation power in mixtures containing xenon [the upper curve in Fig. 5 and curve (*1*) in Fig. 6] can be caused by quenching of the $C^2\Pi_{1/2}$ state accompanied by excitation of the $B^2\Sigma^+_{1/2}$ state:

$$HgI\,(C^2\Pi_{1/2}) + Xe \rightarrow HgI\,(B^2\Sigma^+_{1/2}) + Xe + \Delta\mathcal{E}, \qquad (1)$$

where $\Delta\mathcal{E}$ is the difference in the energies of the $C^2\Pi_{1/2}$ and $B^2\Sigma^+_{1/2}$ states of HgI molecules. This process was first discovered in experiments on photodissociation of mercury diiodide [16, 17]. The arguments in favour of this assumption may be the changes in the ratio of the radiation pulse amplitude and the decrease in the pulse duration (Fig. 7b) at $\lambda = 444$ nm (the system of bands of the $B \to X$ transition in excimer molecules HgI) in the $HgI_2 - Xe - He$ mixture, and also the data on $k_q \tau = 4.5 \times 10^{-18} cm^3$ [16] [k_q is the rate constant of quenching of the C-state of mercury monoiodide molecules, and τ is the lifetime of $HgI(C^2\Pi_{1/2})$ molecules]. This quenching occurs along with excitation of $HgI(B^2\Sigma^+_{1/2})$ molecules [7, 18, 19]:

$$HgI_2 + e \to HgI(B^2\Sigma^+_{1/2}) + I(^2P_{3/2}) + e. \qquad (2)$$

The change in the uniformity of radiation for the excimer lamp with two inner electrodes is caused by an increase in the value of the parameter E/p (E is the electric field strength, and p is the total mixture pressure), which is due to the decrease in the distance between the electrodes (*4*) and the perforated electrode (*3*) (see Fig. 1b). When the parameter E/p is large, the population of the B-state is higher [20], resulting in the increase in the radiation power emitted by the excimer lamp.

4. Conclusions

Our study of the spectral, integrated, and temporal characteristics of coaxial excimer lamps of two types pumped by a barrier discharge has revealed that the radiation emitted by the lamp with one inner electrode at 444 nm is uniform in intensity over the entire surface of the lamp. For the lamp with two inner electrodes, the radiation intensity in the plane containing the two electrodes is 1.5 times higher than the intensity along the normal to this plane. In the mixture with the composition $HgI_2 : Xe : He = 1 : 15 : 2388$ at the total pressure of 162 kPa, the radiation intensity is maximal. The average and pulse radiation powers for this mixture amount to 0.6 and 130 W, respectively. The operation of the HgI excimer lamps at pump-pulse repetition rates up to 12 kHz is stable.

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